Estimates of anthropogenic CO_2 concentration from repeated ocean measurements made decades apart

Over the past 200 years ~50% of the CO_2 released to the atmosphere via the burning of fossil-fuels or changes in land-use ("anthropogenic carbon") has dissolved in the oceans. This carbon sequestration by natural processes has drastically reduced the global warming effect of mankind's CO_2 emissions. However the dissolution of anthropogenic CO_2 in the future ocean is likely to be reduced due to chemical changes associated with higher CO_2 levels and, possibly, due to changes in ocean circulation associated with climate change. Critical scientific issues for prediction of future carbon sequestration and hence future atmospheric CO_2 levels are the mechanisms underlying ocean CO_2 uptake, the regions of the surface ocean that are responsible, and the depth range within the ocean in which the CO_2 is being stored. This information is also critical to understanding the effects of CO_2-induced ocean acidification.

The paper by Tanhua et al (2007) presents results from a 10,000 km Meteor cruise through the North Atlantic in 2004 (M60/5). The cruise was designed expressly with the goal of directly measuring the build-up of oceanic CO_2 over decadal timescales and we therefore returned to the exact locations that had been sampled by a major US-led expedition in 1981. The earlier US expedition was responsible for collecting the very first high-quality set of carbon and closely-related parameters through an entire ocean basin, and provided the best-available reference for detecting such change over the ocean's considerable natural carbon variability.

Based on our new data, we were able to detect the CO_2 increase over the intervening 23 years with remarkable resolution. However we were able to go even further: and used the result as the basis for a fundamentally new approach to estimation of the total amount of anthropogenic CO_2 in the water column (i.e. the "extra" carbon stored over the ~225 years since CO_2 started increasing in the global environment). The new approach is based directly on measurements of carbon and circumvents several long-standing problems with other methods of anthropogenic carbon estimation. In particular, the new data and their interpretation shed new light on the depth ranges of the ocean in which the anthropogenic carbon is being stored.

To test the new approach, we compared our estimates with simultaneously measured distributions of the anthropogenic tracer CFC-12 and with CCl_4. This included a quantitative comparison with fully independent estimates of anthropogenic CO_2 calculated from the CFC-12 data. For the latter, we used a recently-developed approach that accounts for the mixing of waters of different "ages" within the ocean interior. The treatment of mixing has been a major limitation with earlier attempts to use tracers to infer anthropogenic carbon. The level of both qualitative and quantitative

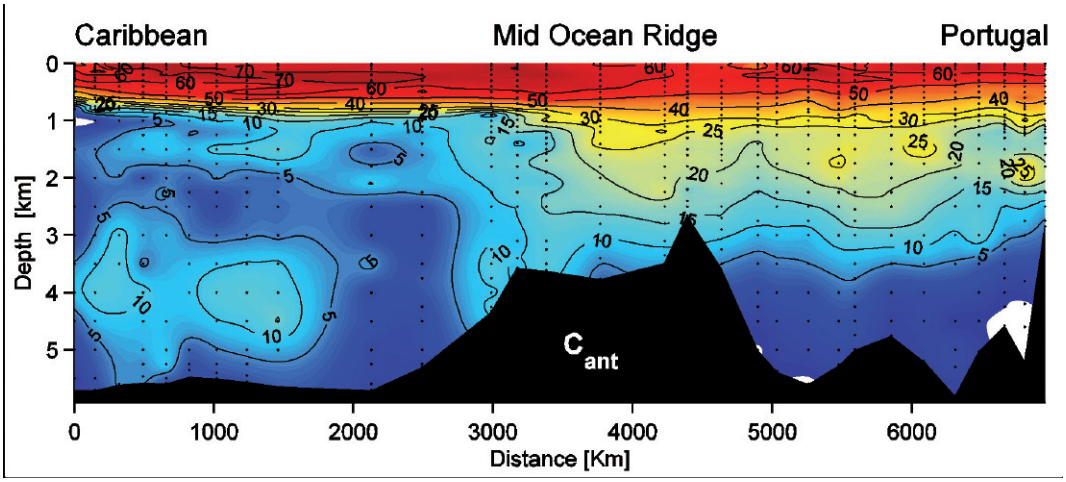

Fig. 1: The content of anthropogenic CO_2 (in µmol kg^{-1}) across the mid-latitude North Atlantic from the Caribbean off Martinique (left) to the coast of Portugal (right) as calculated with the new approach.

agreement between our carbon- and tracer-based approaches is, we believe, unprecedented. We note that, in contrast, nearly every prior comparison of approaches to anthropogenic carbon estimation has revealed large, systematic differences: our result implies that a significant breakthrough has been made.

The overall depth-integrated inventories of anthropogenic carbon were found to be not greatly different between the various approaches which implies that some significant sources of error with earlier approaches cancel each other when averaged over the full depth of the ocean water column. This is encouraging in that it implies that overall global budgets for anthropogenic CO_2 in the Earth system over the past 200 years may be reasonably accurate. (However we note that such "error-cancellation" may not necessarily apply to the critical Southern Ocean, where application of our new approach would now be highly desirable).

Of major significance is the very different depth-distribution of the anthropogenic carbon revealed by our new approach. The depth-distribution of the extra carbon has major implications for projections of future ocean behaviour. If the "extra" carbon is largely stored in the upper ocean and thermocline then future uptake of CO_2 by the ocean will slow down as the chemical buffer capacity of the ocean waters that is upwelled to the surface from intermediate depths is "used-up". On the other hand, storage of a larger proportion of the "extra" CO_2 in the vast, slowly-overturning deep ocean, as revealed by our analysis, implies an extended capacity to store "extra" carbon over longer periods of time.

The deeper input of anthropogenic carbon also has significance for life in the oceans. The preservation and dissolution of calcium carbonate in the oceans, and the ability of carbonate-shell-forming organisms (e.g. deep water corals) to grow in the deep ocean, is dependent on the saturation state of aragonite and calcite at depth. Here one critical issue is whether anthropogenic carbon (and the associated pH change of ocean waters) is impacting the depth and location of the chemical lysocline. Predictions of future change depend on knowledge of where in the ocean the "extra" CO_2 is going: storage above the depth of the lysocline will have little or no effect. Storage near the depth of the present lysocline could drive major changes: the lysocline might shift upwards rapidly. Seen positively, associated dissolution of calcium-carbonate sediments will enhance the ocean's capacity to sequester C_{ant} on long time-scales. On the other hand this will make life more difficult for calcifying marine organisms living in this depth range. Our results reveal significant accumulation of anthropogenic CO_2 at the depth of the aragonite lysocline in the eastern basin of the North Atlantic and closer to the depth of the calcite lysocline in the western basin.

Reference

Tanhua, T., Körtzinger, A., Friis, K., Waugh, D.W. and Wallace, D.W.R., 2007: An estimate of anthropogenic CO_2 inventory from decadal changes in oceanic carbon content. *Proceedings of the National Academy of Science of the United States of America*, **104**(9), 3037-3042.

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